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Palasantzas, G

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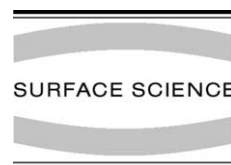


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Adhesion of elastic films on mound rough surfaces

G. Palasantzas

Department of Applied Physics, Materials Science Centre, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands

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Abstract

In this work, we have investigated the adhesive behaviour of elastic films in contact with solid substrates, which are bounded by mound surface roughness. This type of roughness is described by the rms roughness amplitude w , the average mound separation Λ , and the system correlation length ζ . It is shown that both lateral roughness parameters Λ and ζ strongly influence adhesive characteristics. Indeed, with increasing elastic film modulus E , film adhesion is only possible for sufficiently large mound separations Λ . Moreover, the critical elastic modulus E_c (for which spontaneous film decohesion takes place for $E > E_c$) is shown to increase fast with increasing mound separation Λ when $\Lambda \leq \zeta$, while as a function of the system correlation length ζ it increases relatively fast when $\zeta \leq \Lambda$.

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1. Introduction

The influence of surface roughness on the adhesion between an elastic solid in contact with a hard solid substrate is important from the technological and fundamental point of view in systems, which involve polymer/metal junctions. This problem was studied initially by Fuller and Tabor [1], and it was shown that a relatively small surface roughness can remove the adhesion leading to film decohesion. In their model [1] it was considered a Gaussian distribution of asperity heights with all asperities having the same radius of curvature. The contact force was obtained by applying the contact theory by Johnson et al. [2] for each individual asperity considering, however, surface roughness over a single lateral length scale. The maximum

pull-off force was a function of a single parameter which determines the statistically averaged competition between compressive forces from higher asperities that try to pull the surfaces apart, and the adhesive forces from lower asperities that try to hold the surfaces together [1].

Furthermore, random rough surfaces which are commonly encountered in solid surfaces [3,4] possess roughness over many different length scales. This case was considered by Persson and Tosatti [5] for the case random self-affine rough surfaces. It was shown that when the local fractal dimension D is larger than 2.5 the adhesive force may vanish or at least be reduced significantly. Since $D = 3 - H$ with H the roughness exponent which characterises the degree of surface irregularity (as H becomes smaller the surface becomes more irregular at short length scales), the roughness effect becomes prominent for roughness exponents $H < 0.5$ ($D > 2.5$).

E-mail address: g.palasantzas@phys.rug.nl (G. Palasantzas).

So far, the studies with self-affine roughness were limited to roughness exponents $0 < H < 1$, while the case $H = 1$ requires more special attention since it corresponds to a special category of roughness (besides that of the Gaussian roughness as described by the correlation function $\sim e^{-(r/\xi)^{2H}}$ with $H = 1$), which is the mound roughness [6,7]. Indeed, during metal film growth on solid substrates, the growth front can be rough in the sense that multilayer step structures are formed (corresponding also to $H = 1$) [6,7]. In this case the existence of an asymmetric step-edge diffusion barrier inhibits the down-hill diffusion of incoming atoms leading effectively to the creation of multilayer step structures in the form of mounds [6,7]. Examples of mound roughness include the growth of Ag/Ag(1 1 1) by Vrijmoeth et al. [6], the growth of Cu/Cu(1 0 0) by Zuo and Wendelken [6], the growth of Fe/Fe(0 0 1) by Strosio et al. [6] etc. In general, if during roughness formation the corresponding dynamic process leads to a particular wavelength selection, the corresponding morphology can be that of mound roughness.

Therefore, in this work we will address the question of how mound surface roughness influence adhesion properties under conditions of complete and frictionless contact between an elastic film and a solid substrate. It will be shown that the presence of weak roughness can lead to significant reduction of film adhesion and/or film decohesion.

2. Film cohesion theory

We assume that the substrate surface roughness is described by the single valued random roughness function $h(r)$ with r the in-plane position vector $r = (x, y)$ such that $\langle h(r) \rangle = 0$. The adhesive energy is given by [5] $U_{ad} = -\Delta\gamma \int d^2r \sqrt{1 + (\nabla h)^2}$. Upon ensemble average over possible roughness configuration (assuming Gaussian roughness fluctuations) [5] we obtain

$$U_{ad} = -\Delta\gamma A_{\text{flat}} \int_0^{+\infty} dx \sqrt{1 + \rho^2 x} e^{-x},$$

$$\rho = \left[\int q^2 C(q) d^2q \right]^{1/2} \quad (1)$$

with $\rho \left\{ = \sqrt{\langle (\nabla h)^2 \rangle} \right\}$ the average local surface slope, $C(q)$ the Fourier transform of the height correlation function $C(r) = \langle h(r)h(0) \rangle$, A_{flat} the average macroscopic flat contact area, and $-\Delta\gamma$ the change of the local surface free energy upon contact. Further, the elastic energy stored in the elastic film (of elastic modulus E and Poisson ratio ν) is given by [5] $U_{el} = -(1/2) \int d^2r \langle h(r)\sigma_z(r) \rangle$ (assuming the normal displacement field to be $h(r)$). Since $h(q) = [-2(1 - \nu^2)/Eq] \sigma_z(q)$ with $h(q) = (2\pi)^{-2} \times \int h(r) e^{-iqr} d^2r$, we obtain [5]

$$U_{el} = A_{\text{flat}} \frac{E}{4(1 - \nu^2)} \int q C(q) d^2q. \quad (2)$$

The change in the total free energy of the elastic film in contact with the rough substrate is given by $U_{ad} + U_{el} = -A_{\text{flat}} \Delta\gamma_{\text{eff}}$ [5] with effective surface energy $\Delta\gamma_{\text{eff}}$

$$\Delta\gamma_{\text{eff}} = \left(\Delta\gamma \int_0^{+\infty} dx \sqrt{1 + \rho^2 x} e^{-x} - \frac{\pi E}{2(1 - \nu^2)} \int_0^{Q_c} q^2 C(q) dq \right) \quad (3)$$

with $Q_c = \pi/a_o$ and a_o of the order of atomic dimensions. Eq. (3) allows the calculation of the decohesion force by assuming a slab of thickness d that undergoes a displacement \tilde{u} upon the action of a force F_{eff} . The decohesion force is obtained by equating the elastic energy $A_{\text{flat}} d (1/2) E (\tilde{u}/d)^2$ to $A_{\text{flat}} \Delta\gamma_{\text{eff}}$ and taking into account the relation $F_{\text{eff}} = A_{\text{flat}} E (\tilde{u}/d)$ which yields [5,8]

$$F_{\text{eff}} = F_{\text{flat}} (\Delta\gamma_{\text{eff}}/\Delta\gamma)^{1/2} \quad (4)$$

with $F_{\text{flat}} = A_{\text{flat}} (2\Delta\gamma E/d)^{1/2}$. Eq. (4) is valid for constant strain field in the elastic film, which is the case for the planar geometry under consideration [5].

3. Mound roughness model

Mound rough surfaces have been described by the interface roughness amplitude w , the system

correlation length ζ that determines how randomly the mounds are distributed on the surface, and the average mound separation Λ [7]. In Fourier space the mound morphology can be described by the simple model [7]

$$C(q) = \frac{1}{(2\pi)} \frac{w^2 \zeta^2}{2} e^{-(4\pi^2 + q^2 \Lambda^2)(\zeta^2/4\Lambda^2)} I_0(\pi q \zeta^2 / \Lambda) \quad (5)$$

with $I_0(x)$ the modified Bessel function of first kind and zero order. If $\zeta \ll \Lambda$ the roughness reproduces behaviour close to that of Gaussian roughness which corresponds to roughness exponent $H = 1$. For $\zeta \geq \Lambda$, the correlation function $C(r)$ for mound roughness has an oscillatory behaviour leading to a characteristic satellite ring at $q = 2\pi/\Lambda$ for $C(q)$ [7].

4. Results and discussion

The calculations in the following were performed for $\Delta\gamma = 4.8 \times 10^{-2} \text{ J/m}^2$ [5], rms roughness amplitude $w = 1 \text{ nm}$, $a_o = 0.3 \text{ nm}$, and Poisson ratio $\nu = 0.35$. For $E = 0$ (absence of interfacial elastic energy stored in the system) the main roughness contribution for $\Delta\gamma_{\text{eff}}$ comes from the adhesion energy (Fig. 1) and thus from the local surface slope ρ . This is the case of polymer adhe-

sives deposited in liquidlike form on solid surfaces followed by drying. However, shrinkage stress may develop which can diminish the adhesion. With increasing elastic modulus E , the effective energy $\Delta\gamma_{\text{eff}}$ decreases even to values lower than that of flat surfaces ($\Delta\gamma_{\text{eff}} < \Delta\gamma$). However, as the average mound separation Λ increases (leading to surface smoothening) $\Delta\gamma_{\text{eff}}$ approaches values close to $\Delta\gamma$ effectively for $\Lambda > \zeta$ and E in the MPa range. The contribution of the adhesive term on the effective surface energy $\Delta\gamma_{\text{eff}}$ can be further simplified if we calculate the local slope $\rho = [\int_{0 < q < Q_c} q^2 C(q) d^2 q]^{1/2}$ by extending the upper limit of the integration to infinity since in general $Q_c \zeta \gg 1$. In this case we obtain the simple analytic result $\rho \cong 2w(\zeta^{-2} + \pi^2 \Lambda^{-2})^{1/2}$ [7]. Assuming weak roughness or $\rho < 1$ ($w < \zeta, \Lambda$), expansion of Eq. (1) yields the analytic result

$$U_{\text{ad}} \cong -\Delta\gamma_{\text{eff}(E=0)} A_{\text{flat}},$$

$$\Delta\gamma_{\text{eff}(E=0)} = \Delta\gamma \left\{ 1 + 2w^2 \left(\frac{1}{\zeta^2} + \frac{\pi^2}{\Lambda^2} \right) + \sum_{n=2}^{+\infty} R(n) w^{2n} \left(\frac{1}{\zeta^2} + \frac{\pi^2}{\Lambda^2} \right)^n \right\} \quad (6)$$

with $R(n) = \{1 \cdot 3 \cdot 5 \cdots (2n-3)\}(-1)^{n-1} 2^n$.

As Fig. 2a indicates the effect of the system correlation length ζ is rather weak on the value of $\Delta\gamma_{\text{eff}}$, while that of the average mound separation Λ (Fig. 2b) is clearly more significant leading to larger values of E after which spontaneous film decohesion takes place ($\Delta\gamma_{\text{eff}} < 0$). Indeed, for relatively small average mound separation Λ in the nanometer range (and $\Lambda < \zeta$), film decohesion takes place for relatively small elastic modulus E ($< 0.1 \text{ MPa}$). The critical elastic modulus E_c for which $\Delta\gamma_{\text{eff}} = 0$ is given by

$$E_c = 2(1 - \nu^2) \Delta\gamma \int_0^{+\infty} dx \sqrt{(1 + \rho^2 x)} e^{-x} \left/ \left[\pi \int_0^{Q_c} q^2 C(q) dq \right] \right. \quad (7)$$

For elastic modulus $E > E_c$ we have $\Delta\gamma_{\text{eff}} < 0$ leading to spontaneous decohesion (without application

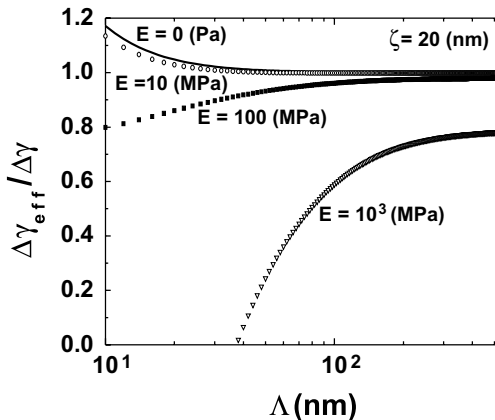


Fig. 1. Effective surface energy $\Delta\gamma_{\text{eff}}$ versus the average mound separation Λ for various elastic modulus E .

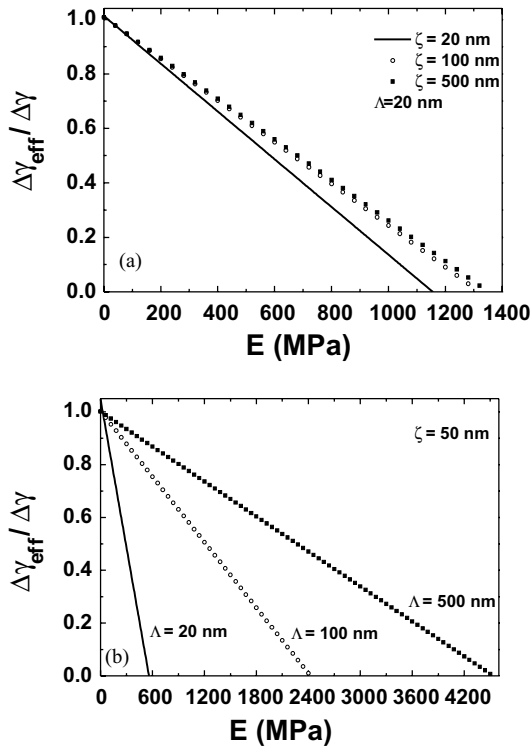


Fig. 2. (a) Effective surface energy $\Delta\gamma_{\text{eff}}$ versus elastic modulus E for various system correlation lengths ζ . (b) Effective surface energy $\Delta\gamma_{\text{eff}}$ versus elastic modulus E for various mound separations Λ .

of any force) of the elastic film, while for $E < E_c$ a finite force will be necessary to decohere the elastic film. Fig. 3a shows that E_c grows fast with average mound separation Λ when $\Lambda \leq 4\zeta$. For $\Lambda \gg \zeta$, the critical elastic modulus E_c evolves rather slowly with further increment of the mound separation Λ . The inverse behaviour is observed in Fig. 3b if one considers E_c as a function of the system correlation length ζ where E_c increases as long as $\zeta \leq \Lambda$.

Furthermore we will calculate the effective decohesion force (Fig. 4). Indeed, Fig. 4a indicates that the effective force to pull-off the film for low elastic modulus E (\ll MPa) decreases with subsequent surface smoothing (increasing lateral length scale Λ and/or ζ) and approaches values close to that for flat surfaces. With increasing elastic modulus the force to decohere the film becomes lower

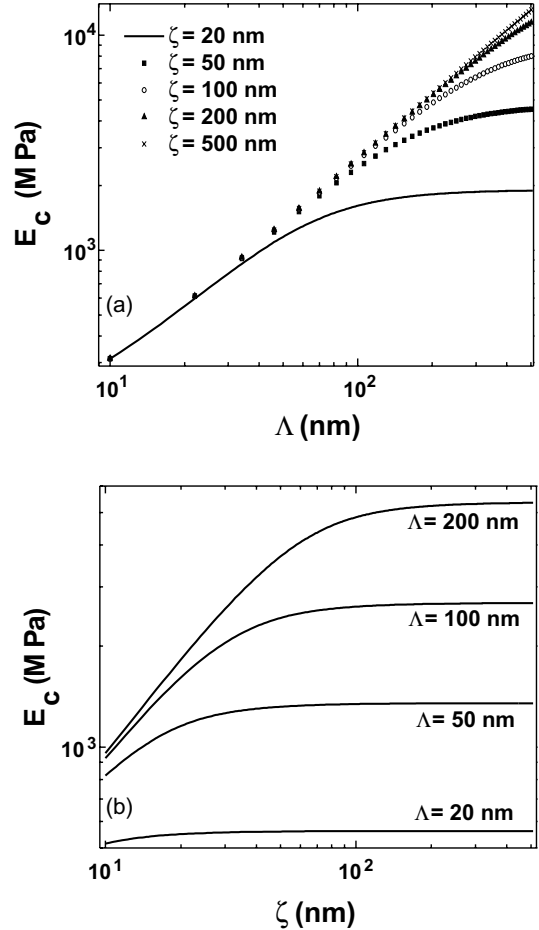


Fig. 3. (a) Critical elastic modulus E_c versus the mound separations Λ for various ζ . (b) Critical elastic modulus E_c versus the system correlation length ζ for various Λ .

for rougher interfaces, which corresponds to decreasing average mound separation Λ (Fig. 4a). Indeed, for $\Lambda < \zeta$ the effective decohesive force decreases significantly in agreement also with Fig. 4b where we plot the effective decohesion force as a function of the elastic modulus E for various values of the mound separation Λ .

The effect of the elastic contribution becomes less significant for $\Lambda > \zeta$ (Figs. 1 and 4a) which corresponds closely to Gaussian roughness. In this case we obtain for the elastic and adhesive energy (upon extension of integration to infinity and assuming $\Lambda > \zeta > w$)

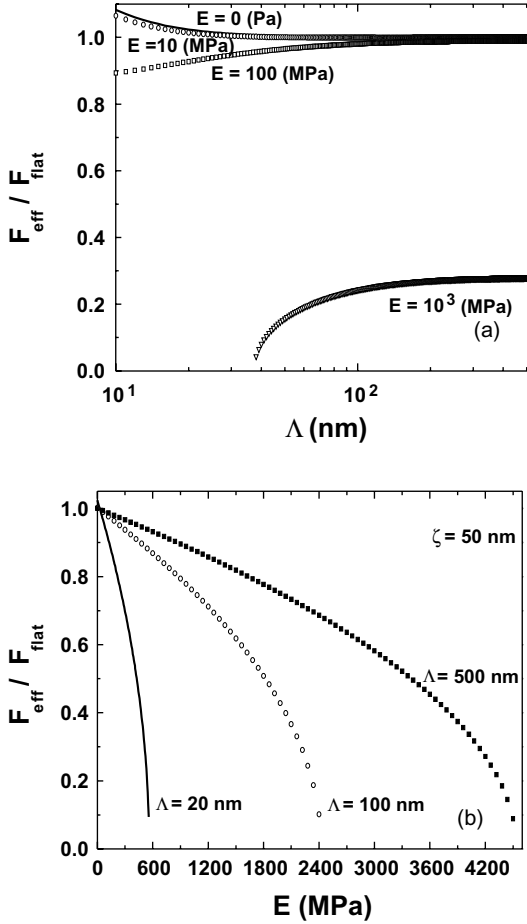


Fig. 4. (a) Effective pull-off force F_{eff} versus the average mound separation Λ for various elastic modulus E , and $\zeta = 20$ nm. (b) Effective pull-off force F_{eff} versus elastic modulus E for various mound separations Λ .

$$U_{\text{el}} \cong A_{\text{flat}} \frac{\sqrt{\pi} E}{4(1-v^2)} \frac{w^2}{\zeta},$$

$$U_{\text{ad}} \cong -\Delta\gamma A_{\text{flat}} \left\{ 1 + 2 \frac{w^2}{\zeta^2} + \sum_{n=2}^{+\infty} R(n) \frac{w^{2n}}{\zeta^{2n}} \right\}. \quad (8)$$

To leading order in w/ζ Eq. (8) yields for $\Delta\gamma_{\text{eff}}$ and E_c

$$\Delta\gamma_{\text{eff}} \cong \Delta\gamma \left\{ 1 + 2 \frac{w^2}{\zeta^2} - \frac{w^2}{\zeta \delta} \right\},$$

$$E_c \cong [4\Delta\gamma(1-v^2)/\sqrt{\pi}](\zeta/w^2) \quad (9)$$

with $\delta = \sqrt{\pi}E/[4\Delta\gamma(1-v^2)]$.

Finally, we should point out that our calculations are strictly limited to the case of elastic films. On the other hand for real polymers [9] viscoelastic effects are present, which alter the value of $\Delta\gamma$ which is considered in the adiabatic limit. In this case modifications are required since surface roughness introduces fluctuating forces with a wide distribution of frequencies [10]. These are effects, which have to be taken into account in order to properly address the adhesive behaviour of viscoelastic films.

5. Conclusions

In summary, we explored aspects of the adhesive behaviour of elastic films in complete frictional contact on solid substrates, which are bounded by mound surface roughness. The latter was described by the rms roughness amplitude w , the average mound separation Λ , and the system correlation length ζ . It is shown that both lateral roughness parameters Λ and ζ could strongly influence adhesive characteristics. Indeed, with increasing elastic film modulus E , film adhesion is only possible for sufficiently large mound separations Λ . Moreover, the critical elastic modulus E_c (for which spontaneous film decohesion occurs for $E > E_c$) is shown to increase fast with increasing mound separation Λ when $\Lambda \leq \zeta$, while as a function of system correlation length ζ it increases relatively fast when $\zeta \leq \Lambda$.

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